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RELATIVISTIC ELECTRON PRECIPITATION, 1: GLOBAL  
ODD NITROGEN CHANGES

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# RELATIVISTIC ELECTRON PRECIPITATION. 1: GLOBAL ODD NITROGEN CHANGES

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*Odd nitrogen produced in the middle atmosphere by reported increases in the particle flux and frequency of occurrence of relativistic electron precipitation from 1981 through 1986 is conservatively calculated to provide an annual source which is as much as 21% of that due to the oxidation of N<sub>2</sub>O. This is a factor of 70 larger than previous estimates for the relativistic electron precipitation source. Our 2-D model calculations, suggests the possibility of significant global scale increases in middle atmospheric NO<sub>x</sub> due to these precipitation events which appear to be associated with solar activity during the declining phase of solar cycle 21.*

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In 1977 and 1980, results were reported<sup>1</sup> suggesting that precipitation of relativistic electrons from the magnetosphere into the middle atmosphere may lead to the formation

of oxides of nitrogen in the lower mesosphere and upper stratosphere and have an impact on the stratospheric ozone layer. Recently, results have been published<sup>2</sup> which suggest that the fluxes and energy levels of relativistic electrons precipitating into the atmosphere during the period from 1981 to 1986 have shown significant increases compared to the period from 1979 to 1980. The observed annually averaged electron flux is shown on figure 1(a). This figure illustrates a seven-fold increase of the precipitating electron flux from the magnetosphere from 1979 to 1984, followed by a decline in 1985 and 1986. The 1986 levels remain a factor of 2.7 above the 1979 levels. Detailed examination<sup>2</sup> of the daily variation of these fluxes reveals a 27-day period. The long-term data record suggests a relationship to the declining phase of solar cycle 21. These measurements were made by the Spectrometer for Energetic Electrons (SEE) aboard the 1979-053 and 1982-019 spacecraft flying at geostationary orbit ( $6.6 R_E$ ).

In the present paper we calculate the magnitude of the lower mesospheric and stratospheric odd nitrogen production associated with these reported relativistic electron precipitation (REP) events and assess the effect of these flux increases on the stratospheric odd nitrogen budget for the 1982–1986 period. Consideration is given to whether or not odd nitrogen produced by these solar-cycle related REP events could be responsible for global scale increases of stratospheric odd nitrogen which have been recently reported<sup>3</sup> as occurring during the 1979 to 1986 period.

## PRODUCTION OF ODD NITROGEN

It is generally accepted that the odd nitrogen present in the stratosphere has as its major source the oxidation of  $N_2O$  through reaction with  $O(^1D)$ <sup>4</sup>. In the present study we

calculate the production (from  $\text{N}_2\text{O}$ ) of odd nitrogen using measurements of temperature,  $\text{O}_3$ ,  $\text{NO}_2$ , and  $\text{H}_2\text{O}$  from the Limb Infrared Monitor of the Stratosphere (LIMS) together with measurements of  $\text{N}_2\text{O}$  by the Stratospheric and Mesospheric Sounder (SAMS). In this calculation, the  $\text{O}(^1\text{D})$  is determined by evaluating the photochemical equilibrium ratio between  $\text{O}(^1\text{D})$  and observed  $\text{O}_3$ . Values of the reaction rates and cross-sections used in these calculations are those reported in the JPL 1985 document <sup>5</sup>. The radiative fields used in the calculation of the required photodissociation rates were determined with the aid of a multiple scattering code <sup>6</sup> with the overhead opacity determined from the LIMS  $\text{O}_3$  and temperature measurements. All calculations were made for average sunlight conditions for the given season and latitude.

To calculate the production of odd nitrogen associated with REP fluxes, we assume that published <sup>2</sup> 1-D transport calculations describing the deposition of energy as the electrons penetrate the atmosphere are valid for the latitude ranges within which the precipitation is thought to occur, 55–72 degrees north and south. We also assume that the precipitation of these particles occurs essentially in a zonally symmetric manner (Baker, 1987, Private Communication). Since these events occur sporadically, it is important to assess the fraction of time that such events occur. An examination of the time history of these fluxes indicates <sup>2</sup> that these events occurred approximately 10% of the time from 1982 to mid-1986. After mid-1986, particle flux observations indicate that the frequency of occurrence and the energy level of these events declined significantly. In calculating the production of odd nitrogen due to the REP events, we draw upon work <sup>7</sup> describing the ion chemistry, initiated by particle precipitation, that leads to the formation of  $\text{NO}$ .

The recent literature<sup>7</sup> suggests that from 1.2 to 2 molecules of odd nitrogen are formed as NO for each ion pair produced during the absorption of the precipitating particles within the mesosphere and stratosphere. For the present calculations we use an odd nitrogen production rate of 1.2 molecules per ion pair formed.

Shown on figure 1(b) are the production of odd nitrogen due to the oxidation of N<sub>2</sub>O for 60 S, 60 N, and the equator as calculated from the LIMS and SAMS data. These profiles are for the May 1979 time period. Also shown is the reported<sup>2</sup> ionization profile associated with a REP event. We note that this profile was calculated for the electron fluxes associated with a rather modest precipitation event measured by the SEE instrument flown aboard the 1979-053 spacecraft in June 1980. Thus, results reported in this paper *should represent a lower limit to odd nitrogen produced* during the 1982-86 period. The results show that in the upper stratosphere (>40 km) the formation of odd nitrogen due to ionization processes associated with these REP events is larger than the high latitude sources of odd nitrogen due to the oxidation of N<sub>2</sub>O. The peak production rate of odd nitrogen by the REP events occurs at 57 km and exceeds the peak N<sub>2</sub>O oxidation formation rate (which occurs at 30 km at the equator) by 75%.

#### ODD NITROGEN TRANSPORT AND CHEMISTRY

Whether or not this additional source of odd nitrogen leads to a substantial buildup during the course of the precipitation events between 1981 and the end of 1986 depends upon subsequent transport of odd nitrogen produced by these REP events between 55 and 72 degrees north and south and above approximately 40 km. If the major transport from these regions occurs upward from the stratosphere to the mesosphere, there is likely to

be little buildup since within the mesosphere, odd nitrogen has a relatively short lifetime against photochemical destruction except in the fall and winter months. For 1979 at the upper stratospheric levels (53 km) for May, the odd nitrogen photochemical lifetimes for 60°N and 60°S are approximately 360 and 820 days, respectively. However, the rate of loss of odd nitrogen at a point is a quadratic function of the local odd nitrogen and an exponential function of the overhead column sum of NO. Odd nitrogen increases locally would shorten the lifetime. Increases of odd nitrogen above the local point would lengthen it. The net effect would be some combination of the two. If the odd nitrogen formed is transported to the lower stratosphere, a buildup would be expected because its lifetime against combined losses due to photochemical destruction and transport out of the stratosphere is 3–4 years. To assess the fate of the ion-produced odd nitrogen within these regions, we have conducted two-dimensional advective transport studies to determine the motion of the air parcels within which the ion-produced odd nitrogen is formed.

The advective flow fields used for this purpose have been diabatically diagnosed with the aid of a contemporary radiative transfer code using climatological CO<sub>2</sub> values and satellite measurements of O<sub>3</sub>, temperature, and H<sub>2</sub>O typical of 1979–1982. These observations were zonally and monthly averaged with a latitudinal and vertical resolution of 10 degrees and 1.5 km, respectively. With the radiative code, the net radiative heating fields were determined on potential temperature surfaces for each of the 12 months. These radiative heating distributions were then used to calculate the zonally averaged horizontal and vertical advective circulation fields. These procedures, the codes used, and the results

have recently been described<sup>8</sup>. Essentially these velocity fields provide the same transport as more conventional advectively driven two-dimensional models operated with zero or very small values of the quasi-horizontal diffusive transport. They provide a valuable diagnostic calculation of the advective transport as a function of time.

The transport calculations were carried out for each of the 12 calendar months for 150 days using 1-day time steps in both hemispheres. The parcel trajectories in the meridional plane were initiated within the altitude-latitude domain (on a grid with a resolution of 0.5 km and 1 degree) where significant ion production of odd nitrogen occurred. Results of the transport simulation averaged for 12 months indicate that after 150 days approximately 85% of the air parcels are retained within the stratosphere and lower mesosphere. The large majority of the parcels moved poleward and downward. A summary of these results for 2 months is shown on figure 2.

Air parcels which move polewards and downwards into the stratosphere may be expected to retain the increased levels of odd nitrogen formed by ionization which occurs during the deposition of precipitating particle energy and the energy due to the absorption of the associated bremsstrahlung radiation<sup>2,7</sup>. If a substantial fraction of the air parcels do move into the lower stratosphere during 1982-1986, it may be expected that they will accumulate and lead to significant increases of high-latitude lower stratospheric odd nitrogen. Some observational results have been reported<sup>3</sup> from the SAGE, SAGE II, LIMS, and SME instruments which suggest that the odd nitrogen levels have increased. As a result of recirculation within the stratosphere<sup>3,8</sup>, rapid quasi-horizontal transport, and in-place odd nitrogen formation, increases in the levels of odd nitrogen in the upper stratosphere

should also be expected as a result of increased REP fluxes. The largest increases should occur at the high latitudes.

Odd nitrogen production by the REP events should occur as parcels are transported within the altitude-latitude region of the energy deposition. To evaluate the possibility of the accumulation of odd nitrogen during transport, we select an elemental tracer volume at the 46 km level and allow it to be advected downward into the stratosphere and at the same time calculate the production of odd nitrogen due to REP events as the parcel moves along the trajectory. The production of odd nitrogen is calculated by assuming that the one-dimensional ionization profile shown in figure 1 applies unchanged for all latitudes within the relatively narrow 55 to 72 degree latitude band for each hemisphere. Therefore, the odd nitrogen production within the parcel as it is advected is determined by the ionization rate at the particular altitude along the trajectory within these latitude bands. When the parcel moves outside of the 55-72 degree latitude band, production is set to zero. We also consider the production of odd nitrogen (due to the oxidation of  $N_2O$ ) and the destruction of odd nitrogen as the parcel moves along the trajectory. These calculations are done by using as initial input to the chemical calculations values of the  $N_2O$  concentrations from the SAMS data, and values of  $O_3$ ,  $NO_2$ ,  $H_2O$ , and  $HNO_3$  from the LIMS data set. Initial values of unmeasured radicals and total odd chlorine were taken from published extended species data<sup>9</sup>. Integration of the chemical equations and the determination of the temperatures, photodissociation rates, overhead opacities, and duration of sunlight along the trajectory were accomplished as described in other work<sup>3</sup>.



The results from this calculation are shown on figure 3. This figure shows panels indicating the altitude and latitude of the parcel as it moves along the trajectory. Also shown is a panel portraying the time variation of total odd nitrogen ( $\text{NO} + \text{NO}_2 + \text{NO}_3 + 2 \times \text{N}_2\text{O}_5 + \text{HNO}_3 + \text{ClNO}_3 + \text{HNO}_4$ ) along the trajectory with and without the effects of the ion-produced odd nitrogen increases. As can be seen, when no effects of the REP events are included, there is little change of the total odd nitrogen along the trajectory due to the oxidation of  $\text{N}_2\text{O}$ . This results from the relatively low  $\text{N}_2\text{O}$  levels at the high altitude and latitudes at which the trajectory is started. With the inclusion of the ion source of odd nitrogen, significant changes in the odd nitrogen occur during the 120 day trajectory. The odd nitrogen increases from 6 to 16 ppbv from day 200 to day 250 at which time the parcel moves out of the latitude range where the estimated odd nitrogen formation from REP events occurs. For this trajectory, the excess odd nitrogen produced will accumulate initially in the lower stratosphere at high latitudes contributing eventually, one would suspect, to a global odd nitrogen increase.

Along these trajectories, the production and loss of  $\text{O}_3$  is also calculated. The results shown on the bottom panel of figure 3 indicate that the  $\text{O}_3$  along the trajectory which includes the ion source of odd nitrogen is reduced by as much as 8% during the 120-day trajectory compared to the trajectory without the ion source of odd nitrogen. The destruction is due to increased levels of catalytic destruction by the well-known odd nitrogen cycle<sup>10</sup>. An analysis of the possible global ozone depletions associated with the REP  $\text{NO}_x$  source is discussed in a companion paper<sup>11</sup> (this issue of *Nature*).

As noted, these trajectory calculations incorporate no quasi-horizontal diffusion to account for rapid meridional transport associated with transient, dissipating planetary waves and neglect the effects of zonally asymmetric constituent motions (chemical eddies). These effects are normally present at high latitudes, and their largest impact occurs in the wintertime at pressures less than 10mb. Based upon a recent 2-D model study<sup>12</sup>, inclusion of such diffusion facilitates the transport of N<sub>2</sub>O from the mid-latitudes along isentropic surfaces towards the polar region resulting in a dilution of the air mass moving downward from the upper stratosphere. Given the overall gradients in total odd nitrogen<sup>9</sup> and the expected high levels of odd nitrogen in the region of REP production, the result of such dilution will be lower percentage increases of odd nitrogen in the descending air mass than shown in the parcel on figure 3. However, the odd nitrogen increases will also be mixed over a broader range of latitude. We will use a global 2-D model that includes the effect of diffusion to study the effect of REP events on the global odd nitrogen budget.

#### ESTIMATED EFFECTS ON GLOBAL ODD NITROGEN BUDGET

We first estimate the total amount of odd nitrogen deposited in the stratosphere by the REP events during the period 1982 to mid-1986 when declines in the flux of these precipitating electrons were observed. Based upon a description of these REP events<sup>2</sup>, it is assumed that production of odd nitrogen in both hemispheres is essentially the same, that the production occurs between 55 and 72 degrees in both hemispheres, and that the events occur 10% of the time. We also assume that only half of the odd nitrogen produced within the lower mesosphere (from 53 to 68 km) is transported into the stratosphere (during the fall and winter seasons) and that no odd nitrogen produced above 68 km is available to

the stratosphere. In light of previous studies,<sup>7,8,13</sup> these last assumptions are considered to be very conservative.

The results of these calculations are presented in Table 1. Under the present assumptions, the integrated stratospheric source of ion-produced odd nitrogen for both hemispheres (below 53 km) is  $1.9 \times 10^{33}$  molecules/year. Calculations (from data and models) of the global production of odd nitrogen due to the oxidation of  $\text{N}_2\text{O}$  fall within the range  $1.8 - 3 \times 10^{34}$  molecules/year<sup>4,14</sup> depending upon the  $\text{O}_3$  and  $\text{N}_2\text{O}$  distributions used. The present calculations of this production using SAMS and LIMS data give a global source of  $2.6 \times 10^{34}$  and 2-D model calculations<sup>14</sup> also yield a production rate of  $2.6 \times 10^{34}$  molecules/year. If we adopt the modelled and presently calculated value of  $2.6 \times 10^{34}$ , the stratospheric production of odd nitrogen due to REP events represents 7.3% of the global source of odd nitrogen due to the oxidation of  $\text{N}_2\text{O}$ . For both hemispheres between the latitudes of 45 and 75 degrees, the oxidation of  $\text{N}_2\text{O}$  provides  $2.2 \times 10^{33}$  molecules/year. Within these latitudes, stratospheric production by REP events is 86% as large as the  $\text{N}_2\text{O}$  source. The presently calculated global stratospheric production of odd nitrogen due to REPs is a factor of 70 larger than the REP source previously reported<sup>4</sup>. However, the previously reported<sup>1,4</sup> global mesospheric and stratospheric source of odd nitrogen due to REP events is estimated to range from  $1.4 \times 10^{33}$  to  $1.4 \times 10^{34}$  molecules/year compared to the presently calculated global value of  $0.71 \times 10^{34}$  for altitudes at and below 68 km. Of paramount importance in the present analysis is the fact that the present production of  $\text{NO}_x$ , due to REP fluxes, peaks at 57 km. This permits effective transport of this predominantly mesospheric produced odd nitrogen to the mid-to-lower stratosphere suggesting

the possibility of a major odd nitrogen buildup. Energy deposition at these surprisingly low altitudes is due to the high energy levels of the observed electron fluxes. Significant fluxes of electrons were observed with energy levels between 3 and 7 MEV.

If the odd nitrogen produced at latitudes between 55 and 72 degrees (north and south) and altitudes between 53 and 68 km during the fall and winter seasons is included with the stratospheric production in these budget calculations, then the global odd nitrogen production due to the REPs and deposited in the stratosphere would represent 21% of the global production due to  $\text{N}_2\text{O}$  oxidation. Given that this ion source of odd nitrogen persists for a period of 4–5 years, and since the lifetime for odd nitrogen in the global stratosphere is 3–4 years, there is a clear possibility for a significant odd nitrogen buildup. This additional source of odd nitrogen appears to be consistent with the reported <sup>3</sup> increases in odd nitrogen suggested by the preliminary comparisons of the Stratospheric Aerosol and Gas Experiment  $\text{NO}_2$  data sets (SAGE and SAGE II) and by the comparisons of the LIMS and SME  $\text{NO}_2$  data sets.

To further investigate the possibility of an odd nitrogen buildup due to the REP events, we have carried out time dependent 2-D photochemical model calculations. The model used for these studies <sup>14</sup> was driven by a prescribed seasonally varying diabatic circulation field. Prescribed variations of  $\text{O}_3$  as calculated by the model were also used in the calculation of the production and loss of total odd nitrogen. Baseline distributions of odd nitrogen without the REP source were calculated and compared with distributions determined with the source included. The odd nitrogen source due to the REP events was included for model latitude bands centered at 57, 66, and 76 degrees (north and south). Flux of odd

nitrogen to levels below 58 km was included only for the fall and winter seasons. This is a conservative approximation. The source and flux terms were determined from the ion pair production curve (below 68 km) of figure 1 and the diagnosed advective circulation fields.

The simulations were carried out for 4.5 years with the REP source on (starting from the unperturbed model odd nitrogen distribution which we take to approximately correspond to early 1981) and continued for another 3.5 years with it off. Results are shown as percentage changes in the odd nitrogen for four periods of the fourth year of the simulation on figure 4. This period would correspond to 1984. The largest percentage increases occur at a given time just above and below 1 mb at latitudes greater than 45 degrees in both hemispheres. At the high latitudes ( $>70$  degrees) at 10 mb, the increases range from 25 to 100%. The smallest increases occur near the equator. The largest absolute increases of odd nitrogen occur at the lower altitudes. Note that the largest percentage changes in odd nitrogen occur during the late fall and early winter in each hemisphere. These are the seasons when the mesosphere-to-stratosphere descent rate is the highest. The results are consistent with the maximum REP  $\text{NO}_x$  production occurring at 57 km as indicated on figure 1. We also note that the late fall and early winter are periods when lower mesospheric odd nitrogen will have the longest photochemical lifetime due to the relatively large zenith angle at high latitudes.

The odd nitrogen distributions shown in figure 4 show the accumulation after a 4-year period including the effects of diffusion and any redistribution associated with the march of seasons as represented in a 2-D model. Figure 3 shows only the effects of REP within a single isolated descending air mass. As noted above, the diffusive effects will

lead to the dilution of the descending air mass with attendant odd nitrogen levels being reduced compared to a descending isolated parcel. Comparison of figures 4 and 3 at 10 mb shows significant differences with the 2-D model results significantly lower than the parcel calculations.

Figure 5 illustrates the nature of the odd nitrogen buildup in terms of a column sum of odd nitrogen. Shown is the difference between year 4 (1984) and baseline odd nitrogen column sums (between 50 and 3 mb) for 4 months. The results indicate that the largest increases occur polewards of 50 degrees latitude in both hemispheres. Little increase is observed between 40 S and 40 N. In each hemisphere, the largest column increases occur in the spring of the year reflecting the accumulation occurring during the descent from the mesosphere in the late fall and winter. This is consistent with the largest percentage increases occurring at the upper stratospheric levels in the late fall of the year and the associated odd nitrogen being transported downward during the next several months. Maximum odd nitrogen column sum increases at 80 degrees latitude are between  $6-8 \times 10^{15}$  molecules/cm<sup>2</sup>, or approximately 50% of the model reference odd nitrogen levels at that latitude. This is in very good agreement with the preliminary high-latitude comparisons of the SAGE, SAGE II, LIMS, and SME NO<sub>2</sub> data sets<sup>3</sup>. It also appears to be consistent with the limited comparisons<sup>15</sup> of odd nitrogen species provided by measurements by the LIMS instrument in 1978 and 1979 and by measurements of the Atmospheric Trace Molecule Spectroscopy (ATMOS) instrument in May 1985. Recently reported<sup>15</sup> comparisons of LIMS and SME NO<sub>2</sub> levels at 10 mb also appear to support the presence of a high-latitude increase in NO<sub>2</sub> between 1978-1979 and 1983-1984. Unfortunately, no global data base is

available which would permit the examination of odd nitrogen on a continuing and self-consistent basis during the entire 1979-1986 period. Given the estimated residence time of 3-4 years for odd nitrogen in the stratosphere, one would expect recovery to occur in a similar period. Our calculation showed that by the end of the eighth year (i.e. three and a half years after turning off the REP source), the odd nitrogen distribution has recovered to within 4% of its original baseline value in the lower stratosphere, and within 1% in the upper stratosphere. Given the measurements of relativistic electron fluxes to date, these calculations would suggest that the atmospheric recovery should occur by the end of 1989. This, however, presupposes no further odd nitrogen perturbations. We note that the end of this recovery period falls near the estimated time of the next solar maximum.

#### CONCLUDING REMARKS

The present analysis appears to confirm the importance of REP production of odd nitrogen to the stratospheric odd nitrogen budget (and by extension, to the stratospheric  $O_3$  budget). The fluxes of precipitating relativistic electrons are apparently related to solar activity and the declining phase of solar cycle 21<sup>2</sup>. We note that the observed annual average electron flux peaked in 1984 and declined monotonically to the 1986 levels. The 1984 flux levels were 7.5 times the observed values in 1979. Despite the declines thereafter, the observed fluxes in 1986 remained at 2.7 times the 1979 values. Based upon these measured levels of REP fluxes, this enhanced production of odd nitrogen would be expected to continue throughout the 1982-1986 period with high latitude stratospheric  $NO_x$  concentrations increasing steadily. This suggests that these fluxes of relativistic

electrons should cause observable and significant increases in high-latitude stratospheric odd nitrogen.

Based upon advective 2-D transport calculations, it further appears that most of the odd nitrogen formed within these regions (presumably in the 50–70 degree latitude bands) would be transported poleward and downward providing the possibility of stratospheric accumulation at higher latitudes. Diagnosed advective descent rates and 2-D model calculations suggest that downward mesospheric-to-stratospheric transport is almost certain to occur during the fall and winter in both hemispheres at latitudes polewards of 40 degrees. It is, therefore, highly likely that a significant fraction of the odd nitrogen formed in the lower mesosphere within these 50–70 degree bands (and below 68 km) would be transported into the mid-to-lower high-latitude stratosphere.

The calculated global and annual odd nitrogen production due to this flux of precipitating relativistic electrons has been conservatively estimated to be as large as 21% of the annual production of odd nitrogen due to the oxidation of  $\text{N}_2\text{O}$  during the 1982–1986 period. Two-dimensional model calculations (with the REP source included) also suggest that global stratospheric odd nitrogen levels should increase with the largest accumulations occurring at the high latitudes during the spring of the year. Given the importance of odd nitrogen to the maintenance of global  $\text{O}_3$  levels, it would be expected that global  $\text{O}_3$  would be altered by the presence of this significant additional source of odd nitrogen. This issue is examined in the following paper <sup>11</sup>.

Finally, we note that previous modeling studies <sup>16</sup> have used global observations of  $\text{HNO}_3$  as a tracer in model studies (accounting for its simple photochemistry) to aid



in diagnosing transport circulation in the lower stratosphere. The present conservative analysis suggests the inclusion of the REP source could increase the high latitude odd nitrogen abundance by as much as 50%. Thus  $\text{HNO}_3$  as measured by the LIMS instrument in 1978 and 1979 may be higher than would be the case in the absence of the REP and other mesospheric sources of odd nitrogen. Comparisons<sup>16,17</sup> of 2-D model-calculated  $\text{HNO}_3$  (with no REP or mesospheric odd nitrogen source) have been made with LIMS measurements. The results suggest that for agreement, a high latitude odd nitrogen source may be required as well as the normal  $\text{N}_2\text{O}$  source. Based upon the present analysis, we suggest that the REP and other mesospheric sources of odd nitrogen may reconcile these model-data differences<sup>16,17</sup>. Also, they may explain the relatively large stratospheric odd nitrogen levels inferred<sup>9</sup> from LIMS data, particularly at the high latitudes in the lower stratosphere. Conclusions from previous analyses may have to be modified pending an evaluation of the effect of REP events and other polar sources of odd nitrogen on the stratospheric constituents measured by the LIMS.

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Table 1. Annual Production of Odd Nitrogen<sup>a</sup>

Source	Magnitude	Reference
N <sub>2</sub> O oxidation with low and high O <sub>3</sub>	$1.8 - 3 \times 10^{34}$	Crutzen and Schmailzl <sup>4</sup>
2-D model N <sub>2</sub> O oxidation	$2.6 \times 10^{34}$	Ko et al. <sup>14</sup>
N <sub>2</sub> O oxidation calculated using LIMS and SAMS data	$2.6 \times 10^{34}$	Present work
N <sub>2</sub> O oxidation in both hemispheres between 45-75°	$0.22 \times 10^{34}$	Present work
Precipitating electrons Stratospheric production	$0.19 \times 10^{34}$	Present work
Precipitating electrons Mesospheric ( < 68 km) and stratospheric production	$0.71 \times 10^{34}$	Present work
Precipitating electrons Stratospheric production with fall and winter mesospheric contribution	$0.55 \times 10^{34}$	Present work

<sup>a</sup>Units are in molecules per year.

## FIGURE CAPTIONS

Figure 1. (a) Annually averaged flux of energetic electrons as measured by the Spectrometer for Energetic Electrons flown aboard the spacecraft 1979-053 and 1982-019 at geostationary orbit,  $6.6 R_E$ . (After Baker et al.<sup>2</sup>.) (b) The production of odd nitrogen due to the oxidation of  $N_2O$  is shown for several latitudes for May of 1979 (long dashed lines). Also shown are estimates of the ion-pair production rates due to relativistic electron precipitation (REP) for 1 and 10% frequency of occurrence. The shaded area shows the range of ion pair production suggested by Thorne<sup>1</sup>. The solid curve shows the recently reported ion pair production profile as calculated by Baker et al.<sup>2</sup> from SEE observations. The odd nitrogen production rate (due to REP fluxes) may be determined by multiplying the ion-pair production rate by  $1.2^8$ .

Figure 2. Advective transport paths followed by tracers are shown for calculations beginning (a) January 15, and (b) September 15 and ending 150 days later. The lines shown illustrate the advective paths followed by the tracers with initial positions on the top, middle, and bottom of the rectangular altitude-latitude domains shown. These domains represent the atmospheric regions (below 55 km) where a portion of the deposition of precipitating relativistic electron energy occurs with the attendant odd nitrogen production. The change from a late winter descending motion to a springtime and summertime ascending motion is evident in the Southern Hemisphere for transport initiated in September. For the January transport calculations, 90% of the tracers were retained within the stratosphere after 150 days. For September, 91%

were retained. All tracer losses were to the mesosphere. April transport calculations (not shown) retained 71% of the tracers, the smallest of the 12 calendar months.

Figure 3. The panels (a) and (b) illustrate the advective path followed by an "air parcel" beginning at 46 km and 60°N on day 200, July 19. During the motion of this parcel, the production and loss of  $O_3$  and  $NO_x$  were calculated. The effects of the  $O_x$ ,  $NO_x$ ,  $Cl_x$ , and  $HO_x$  chemical families are included in this calculation as reported<sup>3</sup> in earlier work. Production of  $NO_x$  from both the oxidation of  $N_2O$  and the effects of the REP events are included. (c) Total odd nitrogen levels are shown along the trajectory both with ( $\square$ ) and without ( $\diamond$ ) the ionic source of  $NO_x$ . (d) Percent change in  $O_3$  along the advective path due to inclusion of the ionic  $NO_x$  source.

Figure 4. Latitude-altitude distributions of percentage changes in total odd nitrogen as calculated by a 2-D model<sup>14</sup> with the REP production of odd nitrogen taken to be 1.2 times the ion pair production rate shown on figure 1. The source was applied in the model as described in the text. The results are shown after 4 years of simulation with the odd nitrogen source due to REP events turned on. The unperturbed distribution of total odd nitrogen is that reported in reference 14 and is due to the oxidation of  $N_2O$  with a small contribution due to lightning in the equatorial troposphere. (a) For January 30. (b) For April 30. (c) For July 29. (d) For October 27.

Figure 5. Change in calculated total odd nitrogen column abundance between 50 mb and 3 mb. The changes shown are the differences (in  $10^{15}$  molecules/cm<sup>2</sup>), as a function of latitude, between the fourth year (after the REP odd nitrogen source was turned on) and the reference 2-D model odd nitrogen distributions. The Southern Hemisphere

is shown as negative latitude. At the high latitudes, the largest changes represent an approximate 50% increase compared to the model reference levels.

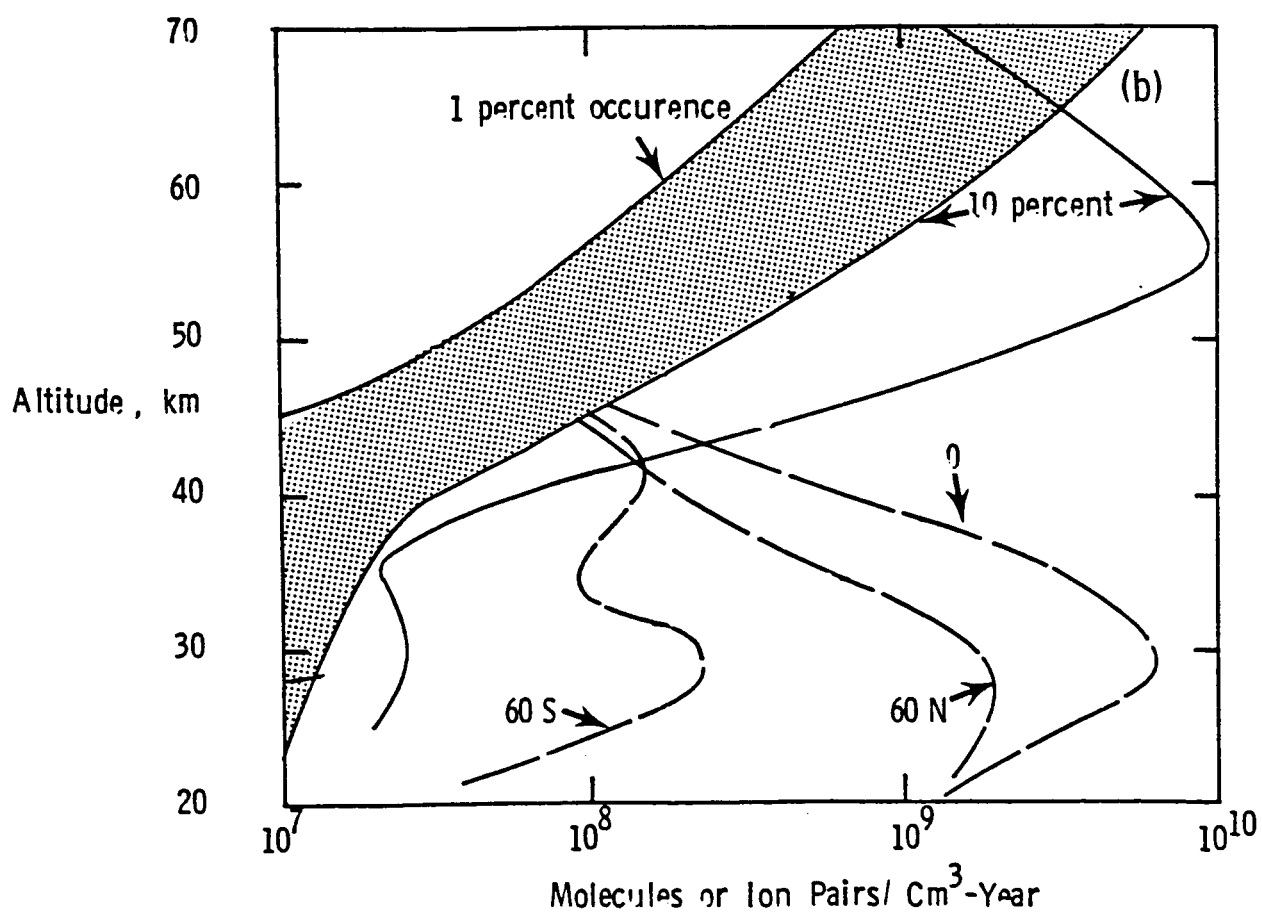
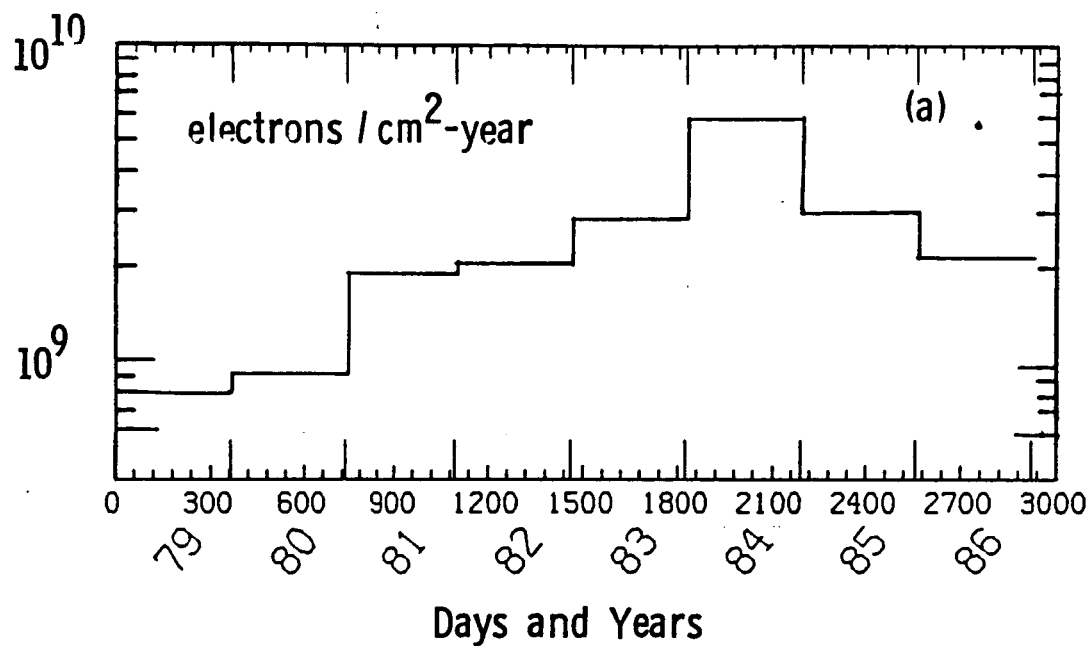


FIG 1



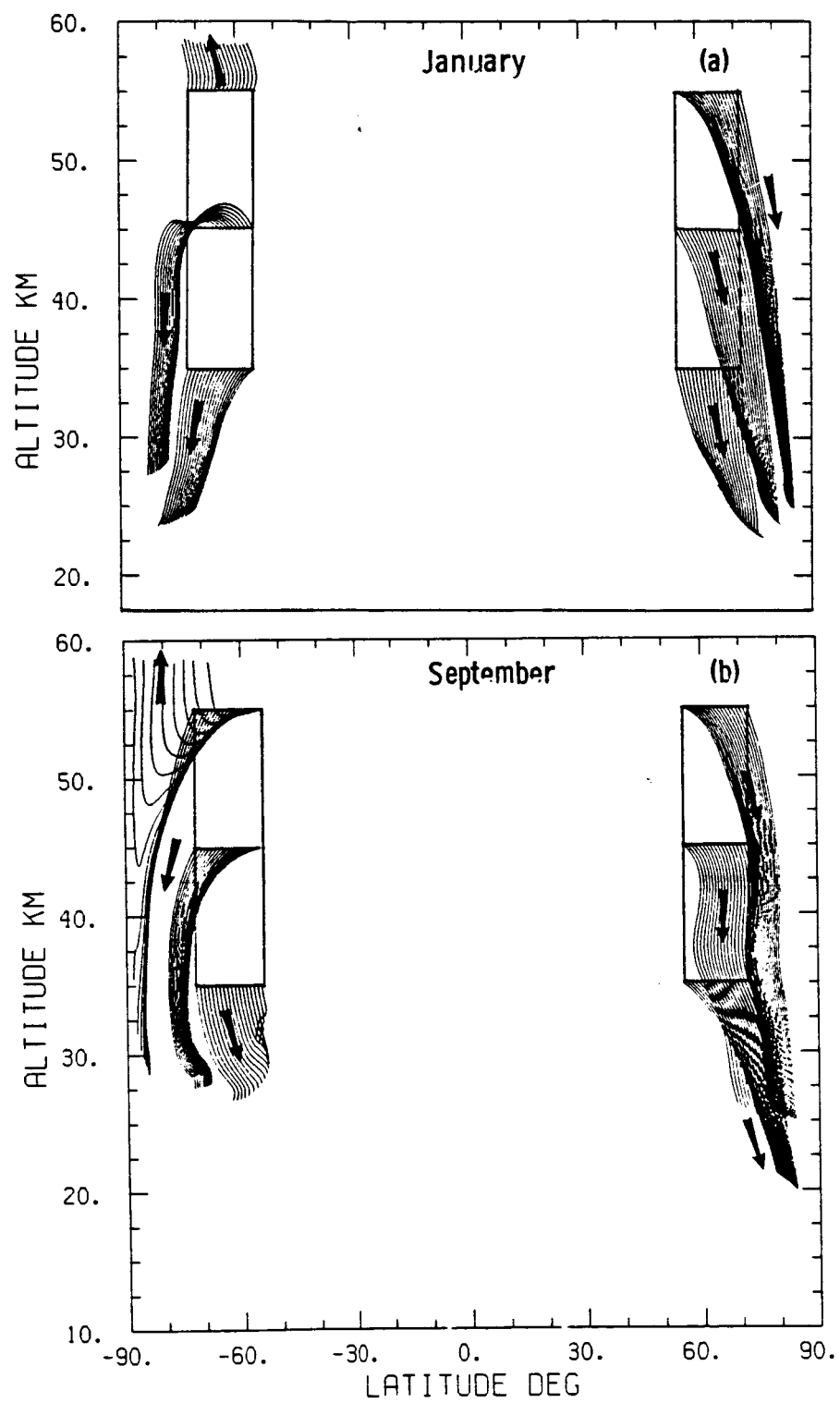


FIG 2

